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Life cycle assessment (LCA) of printed matter: Potential “additives” in recycled paper

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1. Introduction

Concerns about possible effects on human health and the environment from additives/impurities accumulated in globally recycled waste/resources like paper was one of the main reasons for starting up the EU FP7 Coordination Action project RiskCycle (www.wadef.com/projects/riskcycle). A key aim of the project is to identify research needs within this area focusing on both risk assessment (RA) and life cycle assessment (LCA). Besides the sector on paper (being in focus here) also plastics, lubricants, textiles, electronics and leather are included in RiskCycle. In Figure 1 the life cycle of printed matter (paper) is illustrated showing the recycling step which is in special focus in RiskCycle.

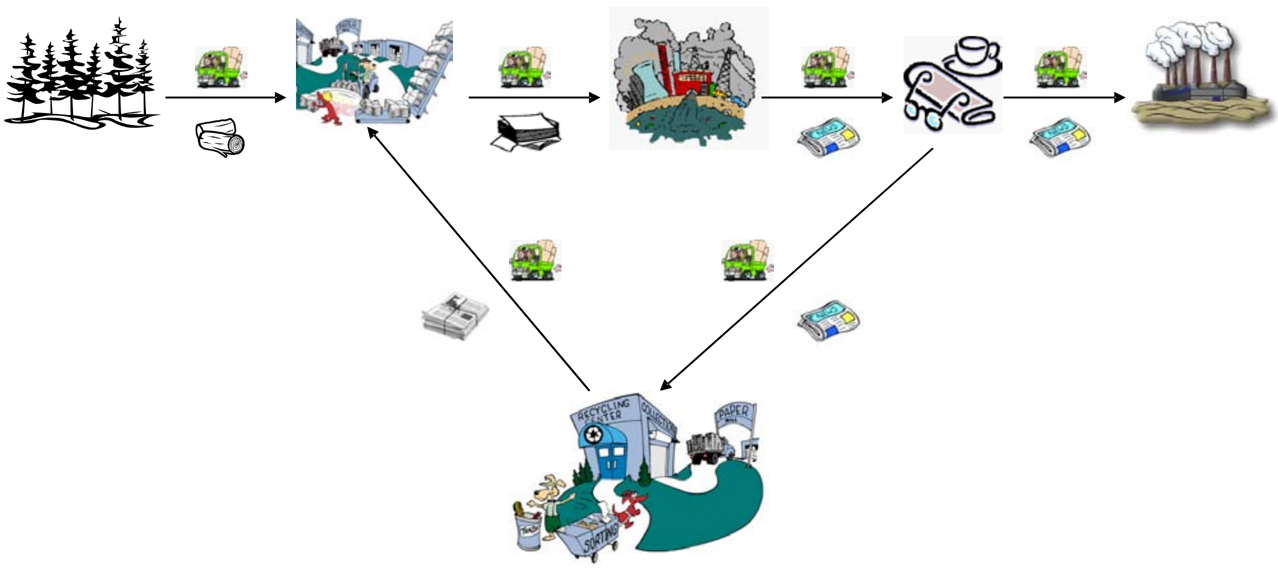

Figure 1: Life cycle of printed matter including recycling [1].

Work package 6 of RiskCycle “Life cycle assessment (LCA) of additives” addresses the issue on how to include additives (including accumulation of additives/impurities in globally recycled waste/resources) in life cycle assessment. Case studies on paper and plastics are going to be performed including the provision of relevant inventory data (process-related resource consumptions and emissions) and life cycle impact assessment (LCIA) characterisation factors for specific additives/impurities. This document deals with the importance of additives/chemicals, used in the printing industry, for the LCA impact profile on printed matter. Furthermore, highly problematic additives/chemicals that might stay/accumulate in the paper when recycled are also addressed. The research reported here is based on an LCA on printed matter [1] and a Danish substitution project [2] – both performed by the author.

2. Life cycle assessment of printed matter

Only a few LCA studies have been done on printed matter (including paper) – mostly focusing on the energy part [1;3]. However, one of the most recent and comprehensive studies [1;3] actually include toxic impacts from chemical emissions – mostly printing chemicals like printing ink of which some components may accumulate in recycled paper. Even though recycling is included in that study there is no special focus on the additives/impurities in the recycled paper. However, the study shows that potential toxic impacts from the production and use of chemicals like pigments, solvents, metals, AOX and biocides may play a very significant role in the impact profile of printed matter as shown below (in brackets: percentage of total normalized and weighted impact potential, EDIP97 methodology):

- Emissions of ink residues (tetradecane) and cleaning agents (hexane, tetradecane) during the printing process and cleaning (35%)
- Emissions (dichlorobenzidine, chloroaniline, cuprous chloride) during pigment production (17-20%)
- Emissions of heavy metals and AOX (as dichloro benzene) during paper production (>3%)
- Emissions of fountain chemicals (i.e. isopropyl alcohol, IPA) during the printing process (6%)
- Emissions of biocides and hydroquinone from the repro- and plate making process (3%)

Anyway, the study only considered a few generic chemical recipes (one printing ink, few cleaning agents etc.) and at least the following shortcomings in need of further research may be identified:

- Ink components (and their precursors) production: siccatives, antioxidants, pigments, dyes etc.
- Water emissions from paper production: softeners (BPA), other phenolic compounds (NPE, APE), other surfactants (LAS), biocides (benzothiazoler, dibromo-compounds), wood extractions (terpenoids, resin acids) and more
- Recycling of paper: Fate of paper chemicals, ink chemicals, glue chemicals etc.
- Treatment of chemical waste: Fate of (hazardous) waste from printing (ink waste, used cleaning agents, used rinsing water etc.) and from recycling of paper (sludge from repulping)

3. Chemicals of high concern in the printing industry

The implementation of the EU REACH regulation will most probably promote substitution within sectors handling a lot of different chemicals like the printing industry. With the aim of being at the cutting edge of this development the Danish printing industry started up a substitution project in 2006. A major part of the work has been mapping the presence of chemicals which are potential candidates for substitution (e.g. PBT, CMR, vPvB, EDS). The mapping comprises a combination of a literature study and an investigation of the actual (2007) presence of candidate substances at 15 Danish printing houses including the examination of almost 900 MSDS's (i.e. products). Furthermore, a focused search in the Danish Product Register has been included.

Table 1. Substances of very high concern (SVHC) appearing on the recently updated EU REACH Annex XIV candidate list and found in the Danish printing industry

Name	CAS No.	Annex XIV criteria	Use
Chromtrioxide	1333-82-0	Carc 1, mut 2	Chrome plating (gravure)
Trichloroethylene	79-01-6	Carc 2	Inks
Cobalt-siccatives *	(10124-43-3)	(Carc 2)	Inks (off-set, screen printing)
Acrylamide	79-06-1	Carc 2, mut 2	Unknown (impurity?)
Pigment Yellow 34 (lead-chromate)	1344-37-2	Rep 1	Inks (screen printing)
Pigment Red 104 (lead-chromate)	12656-85-8	Rep 1	Inks (screen printing)
2-Methoxy ethanol	109-86-4	Rep 2	Photochemistry
Di(2-ethylhexyl)phthalate, DEHP	117-81-7	Rep 2, EDS-list	Inks
Dibutylphthalate, DBT	84-74-2	Rep 2, EDS-list	Inks (screen printing, flexo)
Benzylbutylphthalate, BBT	85-68-7	Rep 2, EDS-list	Inks
Boric acid and borax	10043-35-3 and 1301-96-4	Rep 2	Photochemistry

* Possible content of soluble cobalt(II)salts. Cobalt(II)sulphate, cobalt dichloride, cobalt(II)rbonate, cobalt(II)dinitrate and cobalt(II)diacetate all appears on the recently updated REACH Annex XIV candidate list [25]. IARC classify all soluble cobalt(II)salts as possible carcinogenic, i.e. group 2B (http://monographs.iarc.fr/ENG/Monographs/vol86/mono86.pdf)

More than 200 of the mapped substances are candidates for substitution according to Danish legislation (List of Undesirable Substances) and a total of about 60 of these substances fulfil one or more of the criteria (e.g. CMR, EDS) for the REACH Annex XIV candidate list (Authorisation List).

Table 2. Substances meeting Annex XIV candidate list criteria and found in the Danish printing industry (not listed on the REACH Annex XIV candidate list but potential candidates that may be listed in the future

Name	CAS No.	Annex XIV criteria	Use
Benzene	71-43-2	Carc 1, mut 2	Inks, cleaning agents
Epichlorohydrin	106-89-8	Carc 2	Unknown (impurity?)
2-Methylaziridine	75-55-8	Carc 2	Inks (flexo)
Aziridine	151-56-4	Carc 2, mut 2	Inks (flexo, screen printing)
Propylenoxide	75-56-9	Carc 2, mut 2	Inks, cleaning agents
2-Methoxy propylacetate	70657-70-4	Rep 2	Inks (screen printing)
Triethylene glycol dimethylether	112-49-2	Rep 2	Brake fluid
2-Methoxypropan-1-ol	1589-47-5	Rep 2	Unknown
Alkylphenolethoxylates	(25154-52-3)	EDS-list	Inks, cleaning agents
Chloroalkanes, C14-17	85535-85-9	Possible PBT/vPvB-substance	Chain oil
Octamethylcyclotetrasiloxane (polydimethylsiloxane)	556-67-2 (9016-00-6)	Possible PBT/vPvB-substance	Inks
Bisphenol A	80-05-7	EDS-list	Inks, thermal paper
Resorcinol	108-46-3	EDS-list	Glue
Styrene	100-42-5	EDS-list	Inks, glue
Decamethyl-cyclopentasiloxane	541-02-6	Possible PBT/vPvB-substance	Inks
Stoddard solvent	8052-41-3	Carc 2	Unknown
Solventnaphtha (crude oil), hydrogen treated light naphthen- (benzene >= 0.1%)	92062-15-2	Carc 2	Cleaning agent

In Table 1 and 2 the about 30 substances actually found in the Danish printing industry in 2006 and 2007 (i.e. the novel printing industry inventory and the searches in the Product Register) which meet one or more of the REACH Annex XVI criteria are shown. Eleven of these substances are now (December 2010) part of the Annex XIV candidate list [4], see Table 1. Regarding five out of these eleven substances, i.e. the lead-chromate pigments Pigment Yellow 34 and Pigment Red 104, and the phthalates DEHP, DBT and BBT, inclusion in Annex XIV (Authorization List) is recommended by ECHA and adopted by the Member State Committee [5;6].

Regarding the three phthalates in Table 1, i.e. DEHP; dibutylphthalate, and benzylbutylphthalate, a total yearly consumption above 1 ton, an appearance in about 40 products and a concentration range of 0.1% – 75% in the products are observed in the Danish printing industry. These substances are of interest as they are components of printing inks and remain in the ink after drying and therefore follow the substrate, i.e. paper, plastic or textile, when recycled. They may therefore appear in the recycled material. Actually, according to a German investigation [7] dibutylphthalate have been found in recycled paper used for food packaging. Also other substances in Table 1 and 2 may be of interest as being components of printing inks like the lead chromate pigments, the siloxanes and bisphenol A. Furthermore, 26 hydrocarbon mixtures, most probably containing hazardous single substances (e.g. hexane, heptane, naphthalene) are found in the Danish printing industry. Many of these are used as components in printing inks (and cleaning agents) and therefore may follow the printed substrate when recycled. Some of the hydrocarbon mixtures are used in relatively high amounts in the Danish printing sector like “naphtha (petroleum), hydrodesulfurized (benzene < 0.1%)” used at a total level of 1 500 ton/year, in 35 products with a content of 0.1% – 100%. Finally, it should be noted that highly toxic substances only found in the literature study, like potassium dichromate and hydrocarbon mixtures with high benzene content (>> 0.1%), are probably still in use at places on the world market with less strict environment and health regulation (e.g. Asia), even though phased out on the Danish market. These substances may therefore be relevant when looking at globally recycled printing substrates like paper, plastics and textiles.

4. Conclusions and discussion

Based on the results obtained until now within RiskCycle it may be concluded that in order to perform LCAs on waste/resources recycled globally both new inventory data and new characterisation factors have to be provided. A preliminary solution to the lack of inventory data may be to use Material Flow Analysis and emission factors. One of the main reasons for this lack of useable data on additives for LCA is probably the general focus on energy which has dominated LCA until recently and the lack of consensus on how to include toxicity. Impact categories related to toxicity (and chemicals) are more difficult to handle than e.g. acidification and global warming for which a much higher degree of consensus have existed among method developers for several years. Anyway, consensus on how to deal with human toxicity and ecotoxicity in LCIA is approaching and the USEtox model is probably the best candidate.

The survey of chemicals which are potential candidates for substitution within the Danish printing industry resulted in about 200 substances/substance groups. In total about 60 of these substances fulfil one or more of the criteria for the EU REACH Annex XIV candidate list. Some of these, like the phthalates and the lead chromate pigments, may be relevant when looking at the potential hazard of globally recycled paper based on printed matter.

5. References

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